

REPORT DOCUMENTATION PAGE

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1. REPORT DATE (DD-MM-YYYY)	2. REPORT TYPE New Reprint	3. DATES COVERED (From - To) -		
4. TITLE AND SUBTITLE Raman spectroscopic investigation of boron doped hydrogenated amorphous silicon thin films		5a. CONTRACT NUMBER W911NF-10-1-0410		
		5b. GRANT NUMBER		
		5c. PROGRAM ELEMENT NUMBER 611102		
6. AUTHORS K. Shrestha, V.C. Lopes, A.J. Syllaios, C.L. Littler		5d. PROJECT NUMBER		
		5e. TASK NUMBER		
		5f. WORK UNIT NUMBER		
7. PERFORMING ORGANIZATION NAMES AND ADDRESSES University of North Texas 1155 Union Circle #305250 Denton, TX		8. PERFORMING ORGANIZATION REPORT NUMBER		
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS (ES) U.S. Army Research Office P.O. Box 12211 Research Triangle Park, NC 27709-2211		10. SPONSOR/MONITOR'S ACRONYM(S) ARO		
		11. SPONSOR/MONITOR'S REPORT NUMBER(S) 58295-EL.3		
12. DISTRIBUTION AVAILABILITY STATEMENT Approved for public release; distribution is unlimited.				
13. SUPPLEMENTARY NOTES The views, opinions and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy or decision, unless so designated by other documentation.				
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15. SUBJECT TERMS Amorphous silicon, Plasma enhanced chemical vapor deposition(PECVD), Short- and mid-range order (SRO and MRO), Structural properties, Raman spectroscopy				
16. SECURITY CLASSIFICATION OF: a. REPORT UU		17. LIMITATION OF ABSTRACT UU	15. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON Athanasios Syllaios
b. ABSTRACT UU		c. THIS PAGE UU		19b. TELEPHONE NUMBER 940-565-4561

Report Title

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Continuation for Block 13

ARO Report Number 58295.3-EL
Raman spectroscopic investigation of boron dop...

Block 13: Supplementary Note

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Raman spectroscopic investigation of boron doped hydrogenated amorphous silicon thin films



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ARTICLE INFO

Article history:

Received 31 March 2014

Received in revised form 29 June 2014

Accepted 10 July 2014

Available online 26 July 2014

Keywords:

Amorphous silicon;
Plasma enhanced chemical vapor deposition
(PECVD);
Short- and mid-range order (SRO and MRO);
Structural properties;
Raman spectroscopy

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We report on the effects of hydrogen dilution and boron doping on the structural properties of amorphous silicon thin films prepared by plasma enhanced chemical vapor deposition. Raman studies were performed on samples differing in both boron concentration and hydrogen dilution of the silane precursor prepared at two growth temperatures. Changes in the transverse optical and acoustic phonon modes were analyzed to determine the effect of boron and hydrogen on short- and mid-range order of the amorphous crystal structure. The results show that, with either an increase of hydrogen dilution or growth temperature, the short- and mid-range order improves. However, the effect of growth temperature on the ability of hydrogen to improve the short- and mid-range order decreases as the growth temperature is increased. This is attributed to dissociation of weakly bonded species at higher growth temperature, leading to less hydrogen being absorbed. In addition, for fixed hydrogen dilution, an increase in boron doping results in a decrease in the short-range order due to a rearrangement of the chemical bonding in the thin film. Finally, a direct correlation is seen between the electrical resistivity and the short-range order for samples of differing hydrogen dilution grown at the higher growth temperatures.

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1. Introduction

Raman spectroscopy is one of the most widely used techniques to understand the local crystal structure of amorphous materials since it provides information about the bonding network order in such materials. It is a nondestructive measurement which provides information about the short range order (SRO) and mid-range order (MRO) [1–3]. The SRO of an amorphous material is usually associated with the nearest neighbor coordination [4] and the MRO involves the topology of the network at the level of third or fourth nearest neighbor and described by the size and distribution of rings of constituent atoms [5]. Various groups have used Raman spectroscopy to study the changes in the local crystal structure of a-Si:H by investigating the changes in the transverse optical (TO) and transverse acoustic (TA) modes of vibration [6–9]. These studies show that the intensity ratio of the TA and TO modes (I_{TA}/I_{TO}), full width half maximum of the TO mode (Γ_{TO}), and the central wavenumber (ω_{TO}) of the TO modes depend on growth method, growth temperature, and hydrogen content. In plasma enhanced chemical vapor deposition (PECVD) growth, the structural configuration of a-Si depends on the hydrogen dilution of the silane precursor and the thickness of the thin film, and can yield different states such as amorphous, nano/micro crystalline and polycrystalline phases [10,11]. In addition, structural changes in a-Si:H films due to hydrogen incorporation and doping have been reported [12–15]. The addition of

boron results in local structural changes resulting from the formation of chemical bonds, such as B–Si and B–H–Si, [14] resulting in an increase in bond angle and bond length, leading to a degradation of the SRO. Specifically, it has been shown that hydrogen diluted crystallization is mediated by insertion of H atoms into Si–Si bonds through the formation of a Si–H–Si bonding configuration when H atoms diffuse in a-Si:H matrix, resulting in improved crystallinity, i.e., improvement in SRO [15]. Infrared spectroscopic results [13,16] have shown that, with the addition of boron, there is a competition between boron and silicon to form bonds with the hydrogen, resulting in an increase in B–H stretching mode bonds and a decrease in Si–H bonds as the boron concentration is increased.

In a preliminary work by our group [17], it was observed that an increase of boron doping broadens Γ_{TO} , indicating retardation in the formation of microcrystals in the film, resulting in an increase in structural disorder. It was also shown that higher growth temperature and higher hydrogen dilution promotes improvement in the atomic order of boron-doped a-Si:H films grown by PECVD.

In this work, room temperature Raman spectra from boron doped p-type a-Si:H thin films prepared by PECVD at two growth temperatures for various levels of hydrogen dilution and boron doping will be presented, and the effect of these parameters on the phonon modes of vibration will be discussed. It is found that there is a strong relationship between the local structural order and the growth parameters of temperature, hydrogen dilution and boron doping. The results show that, with either an increase of hydrogen dilution or with increased growth temperature, the SRO and MRO of the a-Si:H films improve, as

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evidenced by a decrease of both Γ_{TO} and the intensity ratio ($I_{\text{TA}}/I_{\text{TO}}$) of the TA and TO vibrational modes [18]. In addition, the effect of hydrogen dilution on SRO and MRO decreases at increased sample growth temperature and, for samples with fixed hydrogen dilution, an increase in boron doping results in a decrease in the SRO. Finally, the electrical resistivity of the samples prepared at the higher growth temperature correlates with Γ_{TO} and ω_{TO} , where the resistivity increases with decreasing Γ_{TO} and increasing ω_{TO} results from the improvement in SRO.

2. Experimental

Several a-Si:H thin films were grown by PECVD at the substrate temperatures of 275 °C and 365 °C. The thickness of the films investigated ranged from approximately 1000 Å to 3000 Å. The flow rate ratio of boron to silane, defined as “ r ” = $[\text{BCl}_3]/[\text{SiH}_4]$, and hydrogen dilution to silane ratio R_H = $[\text{H}_2]/[\text{SiH}_4]$, were varied to produce films with differing hydrogen and boron concentrations. The samples reported here were grown at R_H values ranging from 4 to 55 and with “ r ” values of 0.17 and 0.32. A Thermo Electron Almega XR Raman Spectrometer with a 532 nm laser as an excitation source and a 2.5 micron spot size was used to obtain room temperature Raman spectra. The electrical resistance measurements at room temperature were performed by current voltage (I-V) characteristics in isothermal conditions in the dark. The resistivity was determined from the resistance and thickness of the film.

3. Results and discussion

The Raman spectra for different growth temperatures and hydrogen dilutions for a fixed flow rate ratio of boron $r = 0.32$ are shown in the Fig. (1). In agreement with the growth phase diagram [10], increasing hydrogen dilution for a fixed boron doping and growth temperature results in a decrease in Γ_{TO} , indicating better structural order with an increase of hydrogen dilution. Specifically, Fig. 1(a) shows spectra obtained for a fixed hydrogen dilution of $R_H = 16$ for films grown at 275 °C and 365 °C. It is clearly seen that when either R_H , growth temperature, or both increase, Γ_{TO} narrows, indicating an improvement in SRO. In addition, the intensity ratio, $I_{\text{TA}}/I_{\text{TO}}$, decreases, indicating an improvement in the MRO. The spectra shown in Fig. 1(b) better illustrate the effects of hydrogen dilution on the improvement in local structural order. At the lower growth temperature, an increase in hydrogen concentration results in a substantial improvement in the local (i.e., SRO and MRO) crystalline structure, as evidenced by a decrease of both Γ_{TO} and the intensity ratio ($I_{\text{TA}}/I_{\text{TO}}$) of the TA and TO vibrational modes. This improvement in local order is further supported by the spectra shown in the inset, which shows that at higher hydrogen dilutions, the TO peak shifts to higher wavenumber.

Fig. 1(c) illustrates the effect of growth temperature on the ability of hydrogen to improve the short- and mid-range order of the a-Si:H thin film. This figure shows the spectra obtained for the highest hydrogen dilution of $R_H = 49$ for films grown at 275 °C and 365 °C. At this high hydrogen dilution the Raman spectra obtained at both growth temperatures are virtually identical. This can be understood in terms of changes in the amorphous structure due to bonding nature of silicon with hydrogen. Higher hydrogen dilution affects the growth structure as the kinetics of atom incorporation changes by changing the constituent elements by the formation of either Si–H or Si–H₂. It has been shown by ab initio calculations that mobile hydrogen atoms break strained silicon bonds to form Si–H and Si–H₂ [19]. From the infrared spectroscopy it has been reported that, with the increase of hydrogen dilution, silicon monohydride absorption peaks increase in strength whereas silicon dihydride absorption peaks decrease [13]. For fixed doping and dilution, the silicon monohydride absorption peak is significantly larger for lower growth temperature, [20] which can be attributed to additional hydrogen atoms being absorbed. On the other hand, the higher growth temperature adds surface mobility to the absorbed

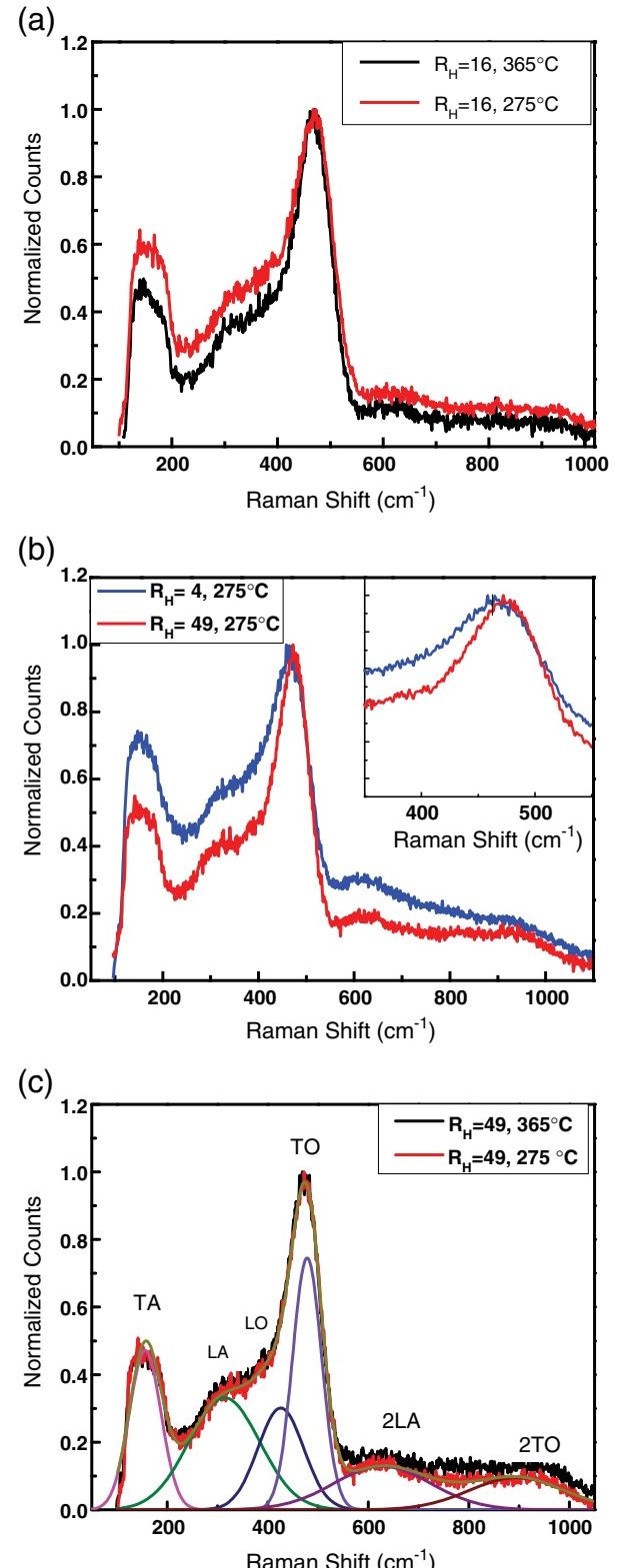


Fig. 1. Representative Raman spectra for the doping level $r = 0.32$ (a) for a fixed hydrogen dilution at growth temperatures 275 °C and 365 °C (b) for the hydrogen dilutions of $R_H = 4$ and 49 at a growth temperature of 275 °C and (c) For the hydrogen dilution ($R_H = 49$) at growth temperatures 275 °C and 365 °C, showing deconvoluted Raman peaks from left to right: TA ($\omega = 155.5 \text{ cm}^{-1}$), LA ($\omega = 312.1 \text{ cm}^{-1}$), LO ($\omega = 425.5 \text{ cm}^{-1}$), TO ($\omega = 477.8 \text{ cm}^{-1}$), 2LA ($\omega = 625.6 \text{ cm}^{-1}$), tentative 2TO ($\omega = 895.8 \text{ cm}^{-1}$).

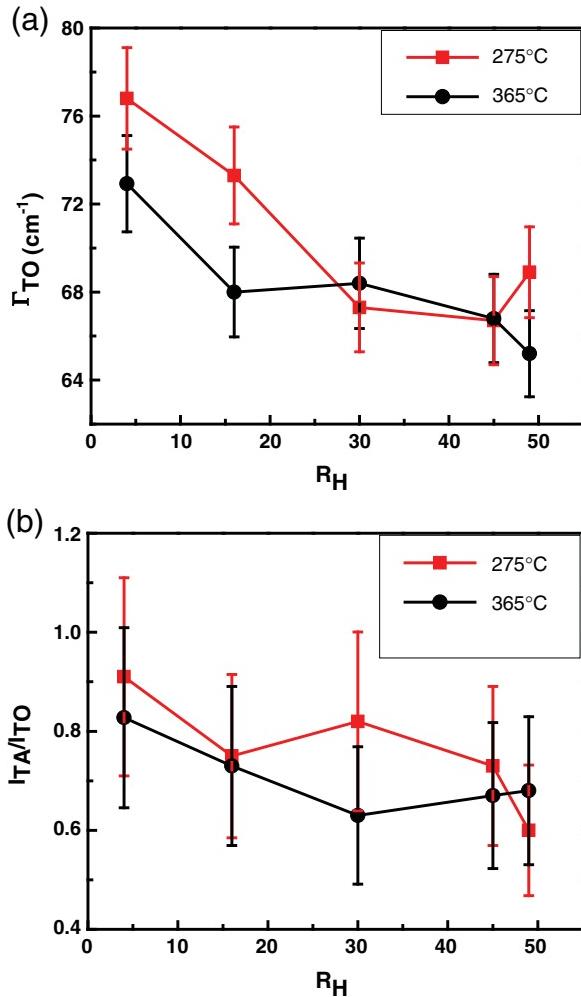


Fig. 2. (a) Γ_{TO} vs. H-dilution, indicating improvement in SRO with increase of H-dilution for “r” = 0.32 (b) intensity ratio (I_{TA}/I_{TO}) vs. H-dilution indicating improvement in MRO with increase of H-dilution for “r” = 0.32.

reactant species, which increases the likelihood of lower energy bond configurations. Further, the higher growth temperature increases the desorption or dissociation of weakly bonded species, reducing the net deposition rate and increasing the likelihood that stronger bond configurations are prevalent. Thus, the improvement in SRO and MRO at each growth temperature due to the formation of Si–H and Si–H₂ bonds overcomes any action of the higher growth temperature to decrease

the absorption of hydrogen. It has been shown in glow discharge grown undoped a-Si:H that mechanical stress is not a primary factor for the change of central frequency position of the TO mode [21]. As a result, it is concluded that the improvement in the local order results primarily from the increase in hydrogen dilution.

As previously discussed, growth temperature and hydrogen dilution cause significant changes in the structural order of a-Si:H thin films. To better quantify the observations shown in Fig. 1(c), Gaussian deconvolution was performed on each spectrum, using 6 Gaussian peaks representing the TA, LA, LO, TO phonon modes and replica peaks of the LA and TO phonon modes, respectively. The results are shown in Fig. (2). Table 1 shows the Raman parameters measured for various samples.

Fig. 2(a) shows Γ_{TO} plotted with respect to the hydrogen dilution ratio (R_H) for both growth temperatures. As the hydrogen dilution is increased, Γ_{TO} decreases, indicating improvement in the SRO. It can be seen from Fig. 2(a) that, the effect of growth temperature on structural order decreases with increasing hydrogen dilution. Fig. 2(b) shows a gradual decrease in the intensity ratio I_{TA}/I_{TO} with an increase of hydrogen dilution, indicating a trend of gradual improvement in the MRO. It can be speculated that the topology of the network improves with the increase of hydrogen dilution [5].

Fig. (3) shows the influence of boron doping on the SRO, illustrated for two hydrogen dilutions (R_H = 40 and 55) and two different boron doping levels (r = 0.17 and 0.32)

As is seen in Fig. 3(a) for both hydrogen dilutions shown, Γ_{TO} increases with increasing boron concentration, indicating degradation in SRO. Illustrated differently, as shown in Fig. 3(b), Γ_{TO} decreases with increasing hydrogen dilution for each boron concentration, indicating that an increase in hydrogen concentration overcomes the effects of boron on the SRO. This suggests that there is a competition between boron and silicon to capture a hydrogen atom and that the degradation in SRO results from a rearrangement of chemical bonding in a-Si:H.

The Raman results show that the structural properties depend strongly on growth temperature and the plasma chemistry. It is well known that hydrogen passivates the dangling bonds of a-Si, [5]. However as seen in Figs. 1(c) and 2(a), increasing the hydrogen supply in the plasma at the higher growth temperature has little effect on the Raman spectra. This indicates that the growth kinetics of PECVD a-Si:H favor hydrogen incorporation into the a-Si thin film at lower growth temperatures. The results presented here agree with and extend those obtained at lower temperatures [18] which showed that the development of the local structure (SRO and MRO) in a-Si:H depends on the initial ordering of the amorphous network which, in turn, is determined by the growth temperature.

Fig. (4) shows a correlation of electrical resistivity with Γ_{TO} and ω_{TO} . In Fig. 4, Γ_{TO} and ω_{TO} are plotted with respect to resistivity for various hydrogen dilutions from 4 to 49 for samples with fixed boron doping

Table 1

Raman parameters (TO frequency, TO line width and intensity ratio I_{TA}/I_{TO}) for measured a-Si:H thin film samples.

Growth temperature (°C)	Thickness (Å)	Boron doping [BCl ₃]/[SiH ₄]	Hydrogen dilution [H ₂]/[SiH ₄]	ω_{TO} (cm ⁻¹)	Γ_{TO} (cm ⁻¹)	I_{TA}/I_{TO}
365	2778	0.32	4	473.1 ± 1.9	72.9 ± 2.6	0.83 ± 0.17
365	3199	0.32	16	474.1 ± 1.9	68 ± 2.4	0.73 ± 0.15
365	3199	0.32	30	477.5 ± 1.9	68.4 ± 2.4	0.63 ± 0.13
365	3162	0.32	45	475.2 ± 1.9	66.8 ± 2.3	0.67 ± 0.13
365	2986	0.32	49	477.2 ± 1.9	65.2 ± 2.3	0.68 ± 0.14
275	3000	0.32	4	473.8 ± 1.9	76.8 ± 2.7	0.91 ± 0.18
275	3000	0.32	16	473.8 ± 1.9	73.3 ± 2.6	0.75 ± 0.15
275	3000	0.32	30	477.0 ± 1.9	67.3 ± 2.4	0.82 ± 0.16
275	3000	0.32	45	475.3 ± 1.9	66.7 ± 2.3	0.73 ± 0.15
275	3000	0.32	49	477.8 ± 1.9	68.9 ± 2.4	0.6 ± 0.12
365	1060	0.32	55	472.0 ± 1.7	74.4 ± 2.2	0.34 ± 0.02
365	1071	0.32	40	471.9 ± 1.7	81.9 ± 2.5	0.44 ± 0.03
365	1092	0.17	55	479.0 ± 1.7	69.3 ± 2.1	0.52 ± 0.04
365	1060	0.17	40	476.5 ± 1.7	74.4 ± 2.2	0.48 ± 0.03

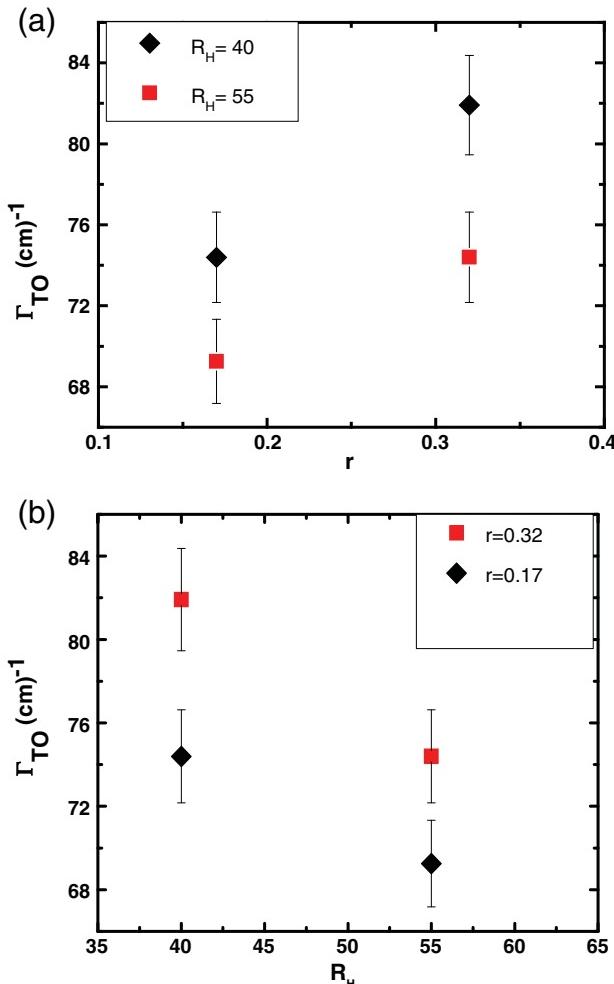


Fig. 3. (a) Γ_{TO} vs. boron incorporation, “r”, and (b) Γ_{TO} vs. hydrogen dilution, R_H .

($r = 0.32$) and growth temperature ($365\text{ }^\circ\text{C}$). The decrease in the linewidth and shift to higher wavenumber of the TO peak with increasing hydrogen dilution indicate that there is a continuous improvement in the structure of the amorphous network on the short range scale. Thus, the increase in resistivity can be understood as a result of the improvement in the local order of the a-Si:H thin film. Similar results in fluorinated a-Si:H prepared by the glow discharge deposition method was reported by relating the central frequency of TO mode with the dark conductivity [22]. It will be interesting to determine if this applies to growth conditions other than hydrogen dilution.

4. Conclusions

The effects of hydrogen dilution and boron doping on the structural properties of amorphous silicon thin films prepared by plasma enhanced chemical vapor deposition are reported and discussed. The results of Raman measurements obtained from a variety of samples of differing hydrogen and boron concentrations grown at two temperatures show that the short- and mid-range order improves with either an increase of hydrogen dilution or higher growth temperature. The effect of growth temperature on the ability of hydrogen to improve the short- and mid-range order decreases as the growth temperature is increased. This is attributed to dissociation of weakly bonded species at

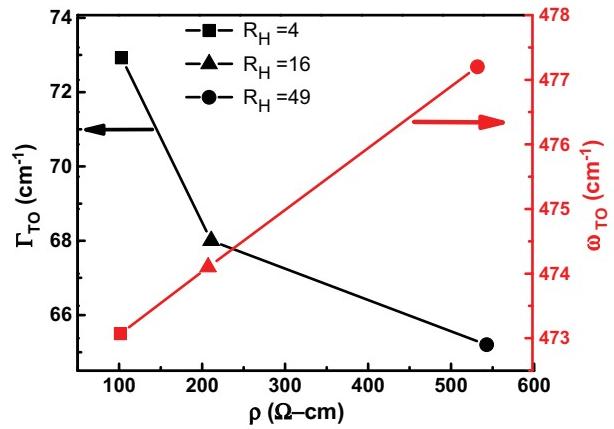


Fig. 4. Line width and central frequency of TO mode vs. RT resistivity for “r” = 0.32, showing improvement in SRO and higher resistivity for higher H-dilution at $365\text{ }^\circ\text{C}$.

higher growth temperature, leading to less hydrogen absorption. For fixed hydrogen dilution, an increase in boron doping results in a decrease in the short-range order, indicating that boron rearranges the a-Si:H matrix either by the appearance of B–H bonds, or Si–H or B–H–Si complex. Finally, a direct correlation is seen between the electrical resistivity and the short-range order for samples of differing hydrogen dilution grown at the higher growth temperatures.

Acknowledgments

This work is supported by ARO grant W911NF-10-0410, William W. Clark Program Manager. The samples were provided by L-3 Communications, Dallas, Texas.

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